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Third Harmonic Terahertz Response Optimization of Doped Monolayer Graphene

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Due to the linear dispersion of graphene, it has recently been predicted that third harmonic generation should be observed in monolayer graphene [1]. Although recent experiments have demonstrated third harmonic generation in a 45-layer thick sample [2], there has been no indication of harmonic generation in monolayer graphene [3]. To understand why this is the case and to try to maximize the third harmonic, it is important to investigate the parameters that are mostly affecting the third harmonic response. In this work, we employ a model that we developed recently [4] based on density-matrix formalism in the length gauge to calculate the nonlinear terahertz (THz) response of doped monolayer graphene and examine the effects of the incident field amplitude, the Fermi level and the scattering time on third harmonic generation. In all cases studied here, there is an optimum Fermi energy for a given scattering time. We consider three different Fermi-energy-dependent scattering mechanisms: phonon, long-range impurity, and short-range impurity. We find that the optimized Fermi energy increases as the scattering time increases. This arises because when the Fermi level is increased, there is an increase in the number of carriers, which in turn leads to a larger current density. In principle, this could lead to a stronger nonlinearity in the transmitted field. However, when the Fermi level is high and the scattering time is too short, only a relatively small fraction of the carriers are driven in k-space by the field to the one side of the Dirac point. As it is the asymmetry in the k-space distribution that yields current clipping, the degree of nonlinearity is limited when the Fermi energy is too high. Therefore, using too high or too low Fermi level will result in a decrease in third harmonic response. We thus find that the Fermi energy and the scattering time of the sample are crucial factors if one is to observe high harmonics in monolayer doped graphene. For a sample that is dominated by phonon scattering, for a field amplitude of 75 kV/cm, we obtain an optimized third harmonic field amplitude that is 1.6% of the amplitude of the transmitted fundamental. Experimentally, this level should be measurable using current detection techniques such as the one reported in Ref. [5] where a dynamic range of 90 dB is reported by measuring 1000 scans that were taken in less than a minute.

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